A New Generation of High-performance, Low-cost Candle Filters

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ABSTRACT

Self-propagating high temperature synthesis (SHS) or combustion synthesis of inorganic materials (CSIM) was used to produce oxide based candle filter elements for hot gas clean-up (HGCU). Material combinations possessing suitable mechanical properties and corrosion resistance were identified and studied within the context of combustion synthesis. These combinations were selected to match the temperature, strength and corrosion resistance requirements of operation in combustion environments, in particular pressurized, fluidized bed combustion (PFBC). Filter permeability and capture efficiency were also examined. Materials were exposed to a simulated corrosive environment for 1000-hour duration. In addition, molding techniques to maximize oxygen diffusion/reaction completion and heating/re-heating cycles have been examined to reduce the cost of fabrication while maximizing simplicity.

INTRODUCTION

Both the Department of Energy and the private sector have extensive ongoing programs for the development of hot gas cleanup (HGCU) methods for coal utilization with emphasis on pressurized fluidized bed combustion (PFBC) and integrated gasification combined cycle (IGCC) equipment. A successful filter will eliminate typical downstream filtration elements such as cyclones, baghouses and/or electrostatic precipitators, giving an overall reduction in pressure loss and equipment cost.

Current production candle filters suffer from long-term reliability problems due to reactions between the tube material/binder and the corrosive IGCC/PFBC environments[1,2]. Current manufacturing/processing technology is somewhat limited in producing corrosion resistant and mechanically tough materials. Using self-propagating high-temperature synthesis (SHS) or combustion synthesis of inorganic materials (CSIM), unique combinations of materials are possible, with suitable properties for the demanding filtration environment. In addition to desirable mechanical properties and corrosion resistance, SHS/CSIM process methods offer manufacturing simplicity, versatility, and cost-effectiveness when compared to other manufacturing techniques. The SHS/CSIM process can easily be tailored to the needs of the PFBC/IGCC and incineration community.

APPROACH

SHS/CSIM is a material processing method that utilizes energy from a highly exothermic reaction to sustain a chemical reaction in a combustion wave[3,4]. The reaction is energy efficient, simple, adaptive to complex shape development, and controllable for selective material properties (i.e. porosity, thermal conductivity, density, pore size, etc.). These characteristics make SHS/CSIM ideal for candle filter element development.

The process is initiated by either locally igniting the powder mixture or heating the mixture to some elevated temperature at which a “thermal explosion” occurs. Either method produces a chemical reaction that is sufficiently exothermic to sustain a combustion wave that converts the reactant powder into the desired product. The speed of the combustion wave typically varies from 0.1 cm/sec to 10 cm/sec. The powder composition and initial conditions (i.e.: geometry, reactive volume, heating rate, etc.) determine the maximum reaction temperature which usually ranges from 1000 deg C to 3000 deg C.

The mechanical and filtration properties of the product are dependent upon the nature of the initial mixture, dispersion of constituents, pre-heat temperature, combustion rate, and other secondary process parameters. By varying the quantity and size of reactants, one can alter the final composition and vary material properties such as bulk density, porosity, and permeability. Another factor controlling the structure of the final product is the combustion mode. As stated above, the process is initiated by either igniting the powder and allowing the combustion wave to propagate through the mixture or by heating the mixture and allowing rapid combustion or “thermal explosion” to occur. The “propagation” mode is a slow process requiring very little energy input to initiate the reaction while the “explosion” mode is a quick transformation of the powder mixture into the final product with uniform properties. The explosion mode is characterized by a near instantaneous, isotropic reaction.

Four major classes of SHS materials are under investigation for use in hot gas clean up. Initial work concentrated on glass binder or traditional SHS/CSIM to produce ceramic oxide materials. It is convenient to think of glass binder materials consisting of large mullite “bricks” held in place by glass mortar. Recent work has focused on other classes of SHS/CSIM materials. The second class of SHS is the glass ceramic material. Consisting of ceramic oxides, this material is similar to the glass binder except the glass reacts with the "bricks” causing some sintering between the neighboring bricks. In general, these materials are more dense and stronger. In addition, the glass ceramics are less prone to creep than the glass binder materials. The third and fourth classes of materials are advanced oxide materials with alumina and mullite dispersed within the material matrix. These advanced oxide materials offer excellent creep resistance above 1093C with good corrosion resistance.

RESULTS

Preliminary results for SHS/CSIM materials are well documented[5-7]. Filtration efficiency in excess of 99.5% was demonstrated for particulate with a mean particle size of 0.1 micron. Corrosion testing was performed by coating the samples with a sodium sulfate solution and exposing the materials to air and water vapor at 850C. Negligible loss in material strength was observed. Permeability was found to meet or exceed the industry standard of 1 foot per minute face velocity per inch of water pressure drop (1fpm/in H20).

Computational chemical analyses and sample material processing were conducted to demonstrate the ability of the SHS/CSIM process to produce materials with the desired end products. To date, over 150 different material combinations have been synthesized, tested and optimized. The "explosion" mode was determined to be the best candidate for application to high volume manufacturing processes with the capability to achieve uniform synthesized product with the desired properties.

Figure 1 depicts a typical temperature profile for SHS/CSIM explosion mode synthesis of a sample in a processing furnace. Both the SHS/CSIM powder temperature and furnace temperature are shown as a function of processing time. The SHS/CSIM temperature lags the furnace temperature up to approximately 900 deg C. At this temperature the combustion synthesis process occurs, causing the sample temperature to rise to approximately 2100 deg C, at a rate of greater than 50 deg C per second. At this point, the SHS/CSIM process is complete and the sample is allowed to cool. By optimizing the SHS reaction, one can produce materials that react in the explosive mode at approximately 400 deg C. This leads to great savings in both processing time and equipment expense as heat input is minimized. Figure 2 is a picture of a typical tube-shape produced using the SHS/CSIM explosion mode.

Current work has focused on the fabrication process. Of particular interest is the diffusion of oxygen into the sample during the reaction. One advantage of the SHS/CSIM technique is the production of the ceramic oxides in-situ. It is possible to produce zirconia, titanium dioxide, magnesium oxide, alumina, mullite and several complex oxides from elemental powders. The advantages of this are twofold. First, the cost of the reactant powder is greatly reduced by using elemental materials. Second, the oxidation of the elemental material provides exothermic energy to complete the SHS/CSIM reaction. It is difficult to provide enough oxygen from other reactants to complete the oxidation process and "black coring" occurs. Thus, the required oxygen must diffuse through the powder from the furnace environment, making rigid, solid molds undesirable. The free flowing nature of the SHS powder simplifies the molding process, as it requires little or no packing pressure.

Three molding techniques have been successfully implemented. All rely upon the pyrolisis of a membrane that defines the surface. The expendable membrane and SHS/CSIM powder are supported by a granular matrix, a high porosity metallic mesh / perforated plate, or a highly porous ceramic. Theses methods allow ample oxygen diffusion while maintaining the geometric tolerances required. In addition, all molding techniques take advantage of reusable parts to drive fabrication costs down.

Another area of interest is particle size distribution. By varying the size of a constituent, say mullite, the porosity and strength of the processed material can be varied. Small particles of metal reactants are desirable as they react completely. Large particles of ingredients like mullite are desirable and somewhat inert, as only the particle's surface reacts and sinters, while the center of the particle retains its initial properties. The use of large particles leads to large pores and high porosity. By introducing smaller particles, the porosity can be controlled.

The porosity and properties of the material can be varied through the reaction temperature. By increasing the reaction temperature or by providing re-heat, one can increase the amount of sintering of the glass materials and decrease the porosity. Figure 3 shows the porosity and material strength for different processing temperatures relative to material processed at 1050 C, where processing temperature is defined as the post reaction furnace temperature.

Figure 4 shows the porosity of a given material as a function of re-heat temperature. In addition to allowing for accurate control of the porosity, this data give some insight into the maximum continuous operating temperature of the material. Typically, the material can operate continuously at 85% of the softening temperature, as defined by a sudden drop in porosity due to glass filling the pore structure. In the case of the material shown above, the operating temperature is approximately 980 deg C.

Glass ceramic material samples were exposed to a simulated combustion environment at 880C for 1000 hours. The gas contained 10 ppm (vol) NaCl, 3300 ppm (vol) SO_2, 1100 ppm (vol) HCl, 15% H_2O, 30% O_2, and the balance was N_2. This mixture is fairly representative of an oxidizing coal combustion atmosphere. The exposed glass-ceramic materials showed a slight drop in room temperature strength. The material showed little change in structure or filtration properties. SEM and EDAX results indicate only minor surface reactions occurred between the corrosive gas and the material. In addition, this test was somewhat extreme as most IGCC and PFBC reactors are anticipated to operate at approximately 732C. It is believed that long-term exposure this temperature will have little effect on material properties.

CONCLUSION

A number of different materials have been produced and tested for use in HTHP (high temperature, high-pressure) filtration application using SHS/CSIM fabrication techniques. Compositions can be categorized into four classes including glass bonded, glass-ceramic and advanced material combinations. Materials fabricated using SHS/CSIM have demonstrated desired mechanical properties, corrosion resistance and filtration capabilities. Specific molding techniques have been developed to produce net shapes with the desired surface finish for filtration applications without additional handling. Properties are easily tailored to a specific application by varying particle size distribution, processing temperature, or re-heat conditions. Simplified molding and use of elemental powders to produce complex oxides greatly reduces cost.

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Figure 1 Typical SHS/CSIM Temperature Profile

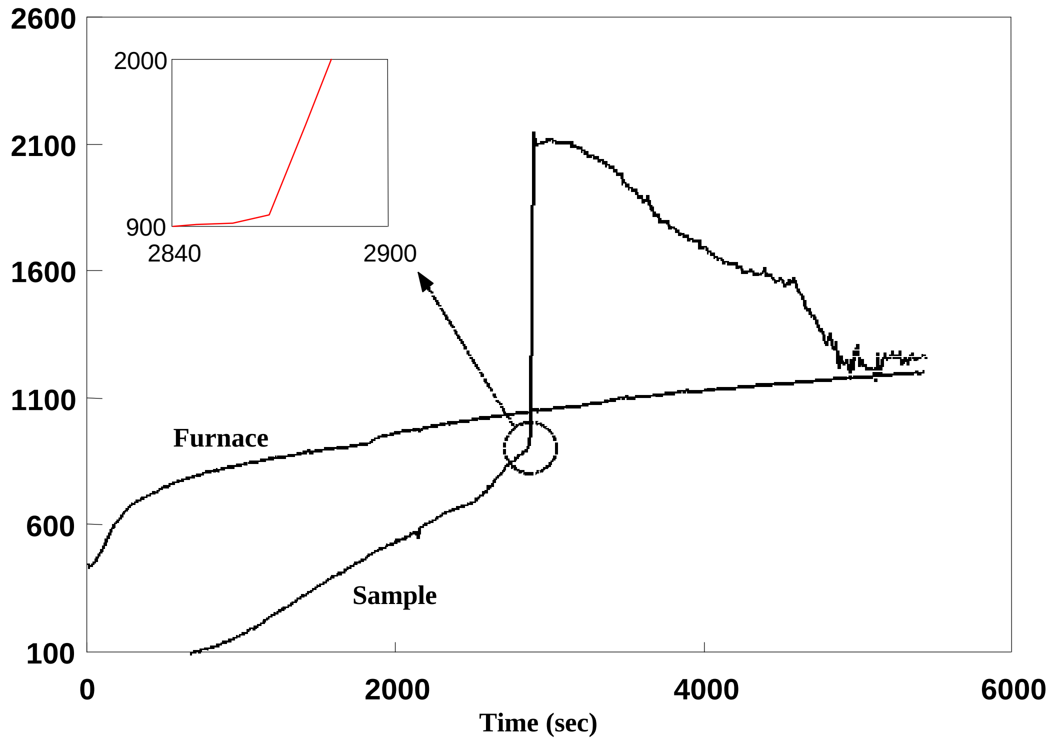

Figure 2 SHS/CSIM explosion mode sample

Figure 3 Effect of Processing Temperature on Material Properties

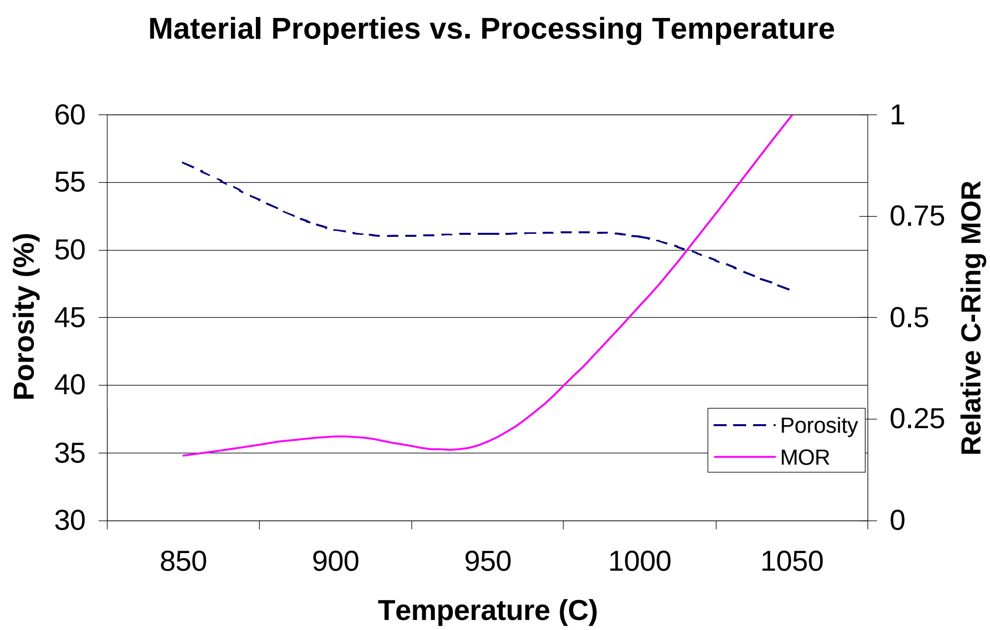

Figure 4 Porosity vs. Re-heat Temperature